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(54) Title: ELECTROLUMINESCENCE IN LIGHT EMITTING POLYMERS FEATURING DEAGGREGATED POLYMERS

(57) Abstract

In general terms, the present invention includes a light emitting polymeric material, the light emitting polymeric material capable of producing electroluminescence upon being provided with a flow of electrons, the light emitting polymeric material comprising a plurality of polymeric chains comprising polymeric chains each having substituent moieties of sufficient number and size and extending from the polymeric chain and about a substantial portion of the circumference about the polymer chain so as to maintain the polymeric chains in a sufficiently deaggregated state (referred to herein as a "strapped" polymer), so as to substantially prevent the redshifting of the electroluminescence and the lowering of light emission efficiency of the electroluminescence.

Electroluminescence in Light Emitting Polymers Featuring Deaggregated Polymers

Technical Field

The present invention is in the field of light-emitting polymers and light emitting devices produced therefrom.

Background

Conjugated polymer based light-emitting devices have become an extensive area of academic and industrial research since the report of electroluminescence (EL) in poly(phenylene vinylene) (PPV) in 1990 [1].

10 A great number of different conjugated polymers have been found to exhibit EL including PPVs [1-3], poly(phenylphenylene vinylene) [4], polyphenylenes [5-7], polythiophenes [8-9], polyquinolines [10], polypyridines [11-12], poly(pyridyl vinylenes) [12-14] and many copolymers of these materials.

15 In addition to many different materials, numerous configurations have been used to change and improve device performance. For instance, the use of additional layers to improve device efficiency



has been known for some time [2,15]. Inserting a hole-transport (electron blocking) layer between the anode and emitting polymer or an electron-transport (hole-blocking) layer between the cathode and emitting polymer can greatly improve efficiency by confining the majority carrier to the emitting layer. A well known hole-transport (electron blocking) layer is poly(vinyl carbazole) (PVK) which has a large band gap (3.5 eV) and is itself luminescent [16-18].

Despite these advances there remains a need for improvements in the electroluminescence performance of light emitting polymers. Particularly, there remains a need to improve the performance of exiplex-forming bilayer devices so as to reduce or eliminate the redshifting believed to be associated with the aggregation of polymeric chains within the emitting polymer.

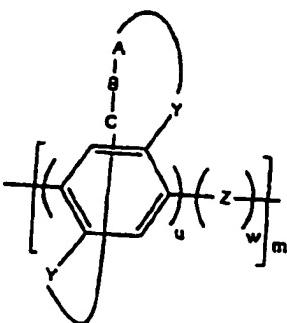
It is a goal of the present invention to produce light emitting polymer and light emitting polymer devices made which give light emissions having reduced redshifting.

In view of the present disclosure or through practice of the present invention, other advantages may become apparent.

Summary of the Invention

In general terms, the present invention includes a light emitting polymeric material the light emitting polymeric material capable of producing electroluminescence upon being provided with a flow of electrons, the light emitting polymeric material comprising a plurality of polymeric chains comprising polymeric chains each having substituent moieties of sufficient number and size and extending from the polymeric chain and about a substantial portion of the circumference about the polymer chain so as to maintain the polymeric chains in a sufficiently deaggregated state (referred to herein as a "strapped" polymer), so as to substantially prevent the redshifting of the electroluminescence and the lowering of light emission efficiency of the electroluminescence.

It is preferred that the polymer of the present invention comprises polymeric chains selected from the group consisting of alternating and random copolymers, having comprising the structure:



10

wherein m is the degree of polymerization;

Y is selected from the group consisting of -CH₂, O, S, CO and NR, wherein R is an alkyl group containing 1 to 16 carbon atoms;

A is selected from the group consisting of (CH₂)_n, (CH₂CH₂O)_n, (CH₂CH₂O)_nNR wherein R is an alkyl group containing 1 to 16 carbon atoms, and aryl groups having 6 to 14 carbon atoms;

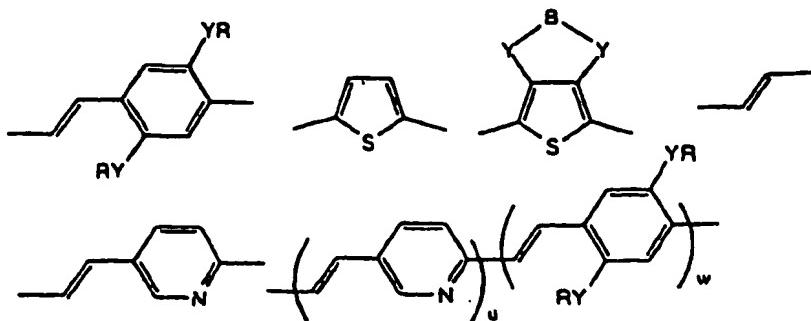
B is selected from the group consisting of (CH₂)_n, aryl groups having 6 to 14 carbon atoms, and calixarene having 18 to 200 carbon atoms;

wherein u may be of a value independently selected from the group 1 to 6, inclusive;

wherein w may be of a value independently selected from the group 1 to 6, inclusive;

wherein n may be of a value independently selected from the group 0 to 6, inclusive; and

wherein Z may be a structure selected from the group consisting of:



5

wherein R is an alkyl group containing 1 to 16 carbon atoms,

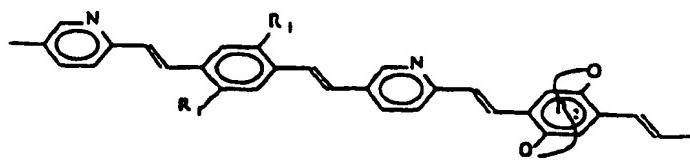
wherein Y is selected from the group consisting of $-CH_2$, O, S, CO and NR_2 wherein R is an alkyl group containing 1 to 16 carbon atoms;

B is selected from the group consisting of $(CH_2)_n$, aryl groups having 6 to 14 carbon atoms, and calixarene having 18 to 200 carbon atoms;

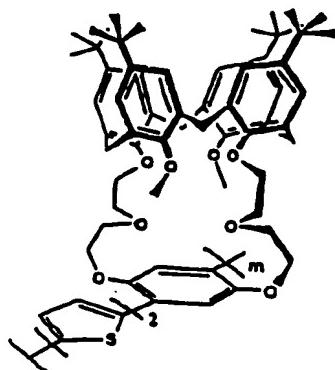
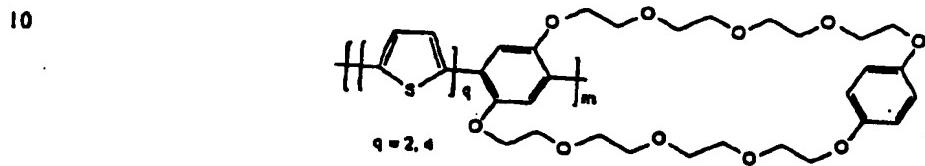
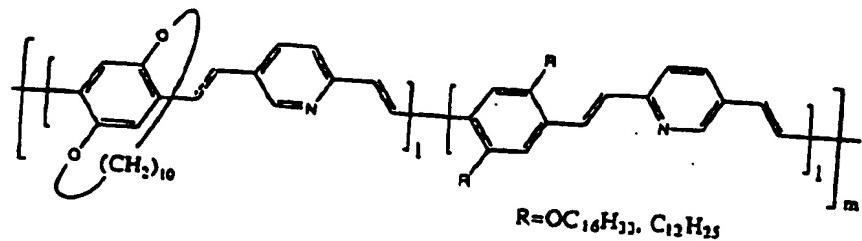
wherein u may be of a value independently selected from the group 1 to 6, inclusive; and

wherein w may be of a value independently selected from the group 1 to 6, inclusive.

15 The electron transporting polymer may include polymeric chains selected from copolymers having the structure:



Some specific examples of polymers used in the present invention include:



There is also provided by the present invention a light emitting polymeric material, said light emitting polymeric material capable of producing electroluminescence upon being provided with a flow of electrons, said light emitting polymeric material comprising:

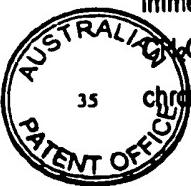
5 a plurality of polymeric chains comprising polymeric chains each being provided with rotaxanes of sufficient number and size and extending from said polymeric chain and about a substantial portion of the circumference about said polymer chain so as to maintain said polymeric chains in a sufficiently deaggregated state, so as to substantially prevent the redshifting of said electroluminescence and the lowering of light emission efficiency of said electroluminescence.

10 The light emitting polymeric material may also be used in single layer, bilayer or other multiple layer devices, using the polymeric material of the present invention. In the case of a single polymeric layer device, the polymeric material of the present invention may be used as the electron transporting/electron blocking layer. In the case of a bilayer or multi-layer devices, the polymeric material of the present invention may be used as the electron transporting layer in conjunction with an electron blocking layer of another appropriate polymer, such as might be selected from the group consisting of poly(vinylcarbazole).

15 The present invention also includes a light emitting device comprising a light emitting polymeric material according to the present invention in all of its embodiments, and a source of electrical current so as to supply the light emitting device with a flow of electrons capable of producing electroluminescence from the device.

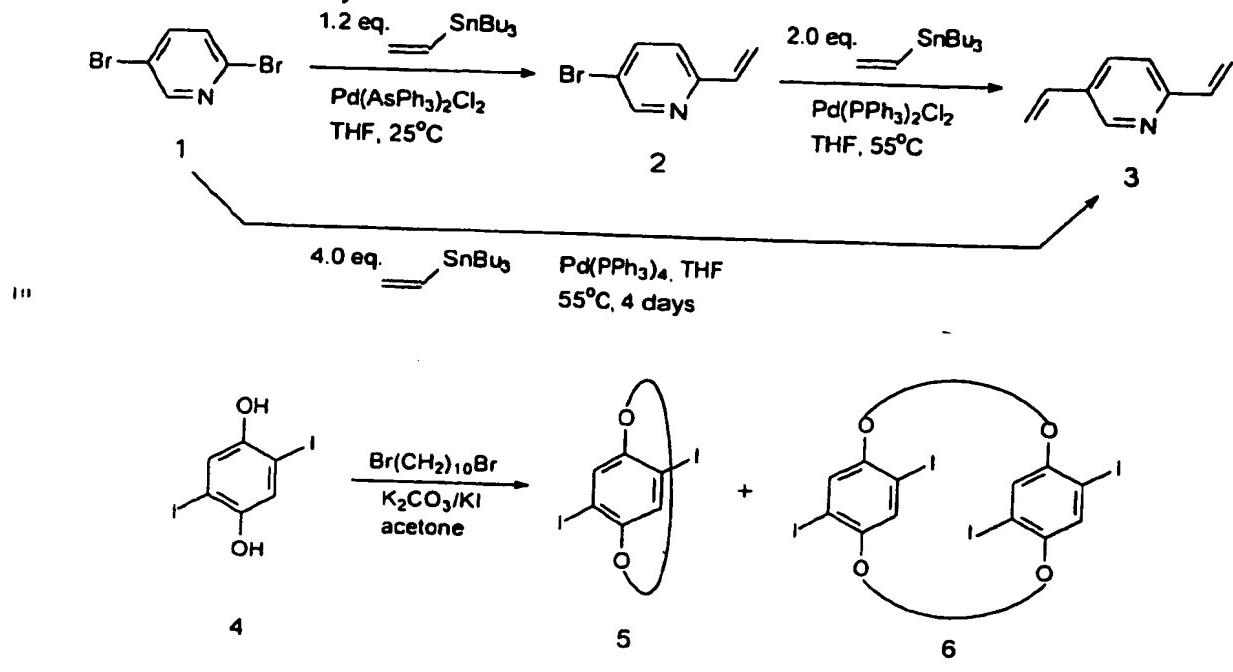
20 Other examples of synthetic methods are described in reference 22 and as follows.

The syntheses of 2,5-divinylpyridine and the macrocyclic monomers are described in Scheme 1. We previously found that by using $Pd(AsPh_3)_2Cl_2$ as a catalyst 2,5-dibromopyridine (1) reacted with vinyltributyltin selectively at the more reactive 2-position to give 5-bromo-2-vinylpyridine (2) in 85% yield. Further reaction with 2 at the 5-position using $Pd(PPh_3)_2Cl_2$ occurs at an elevated temperature to produce 2,5 divinylpyridine (3) in 50% yield. This two-step procedure was replaced by a one-step reaction catalyzed by $Pd(PPh_3)_4$ in the later part of our studies. While the reaction requires prolonged heating (55°C, 6 days) and an excess of vinyltributyltin (4 eq.), it offers a simplified purification procedure and an improved yield of 80% (a 43% overall yield was obtained for the two step procedure). Purification of 3 requires first a distillation under reduced pressure, to give a mixture of vinyltributyltin, 3, and a trace amount of 2. Further purification of the resulting mixture by flash column chromatography gave 3 as clear liquid in high purity (>99.5% by NMR). Monomer 3 was polymerized with the diiodide monomers under Heck reaction conditions immediately after purification in order to avoid free radical polymerization of the vinyl group. Dilute $NaCl_2$ solutions of 3 could be stored for a period of one month in the dark at 0°C and were chromatographed before use. The



6a

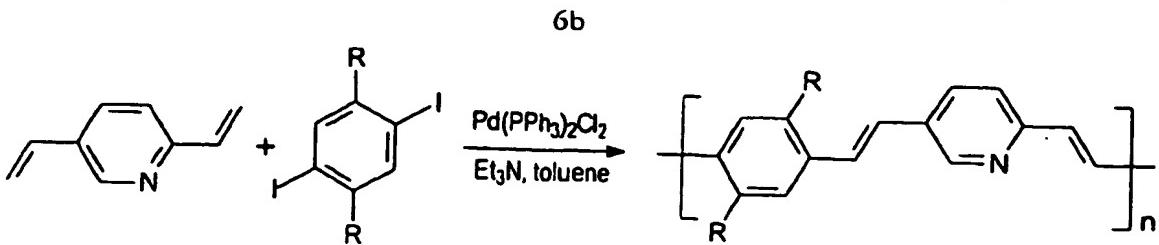
1,4-(oxydecanoxy)-2,5-diiodobezene macrocyclic monomer (5) was easily prepared by a Williamson ether synthesis. In this case, a 1:1 mixture of 2,5-diido-1,4-dihydroquinone (4) and the commercially available 1,10-dibromodecane were slowly added to a suspension of K_2CO_3 and KI in acetone via a syringe pump over a period of three days. The high dilution conditions were designed to maximize the formation of 5, however, a substantial amount (15%) of the undesired adduct, 6, was also produced. Since 5 and 6 were similar in physical properties, their separation was only possible through tedious gravity column chromatography (CH_2Cl_2 /hexane (5:95), SiO_2). Compound 5, was the first fraction and isolated in 25% yield. The structures of 5 and 6 were confirmed by 1H and ^{13}C NMR in conjunction with mass spectrometry.



Scheme 1

As shown in Scheme 2, we have employed Heck coupling reactions for the copolymer syntheses. For the purpose of fine-tuning the band structures of the non-macrocyclic polymers, we synthesised polymers containing disubstituted phenylene units with different electron donating (or withdrawing) abilities. Monomers 7a, 7b and 7c were readily synthesized via standard organic transformations. The polymerizations to form polymers 8a, 8b, and 8c were conducted in toluene at 110°C using $Pd(PPh_3)_2Cl_2$ as a catalyst. In all cases, the precipitation of Et_3N^+ was observed during the polymerization. The precipitation of polymers 8b and 8c was also apparent after 24 hours. To circumvent these problems and improve the efficiency of the coupling reactions, we used modified conditions for the preparation of polymers 9a and 9b. These conditions included a mixed solvent system consisting of DMF and xylene (1:1), for improved solubility, and $Pd(P(o-tolyl))_2Cl_2$ which appeared to be a more active catalyst.





3

7a, R = OC₁₆H₃₃

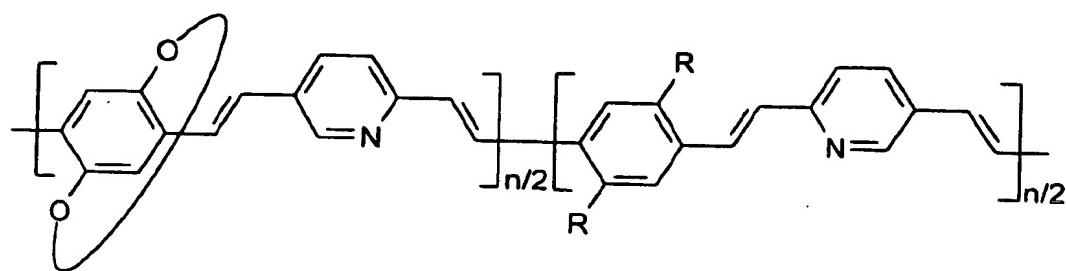
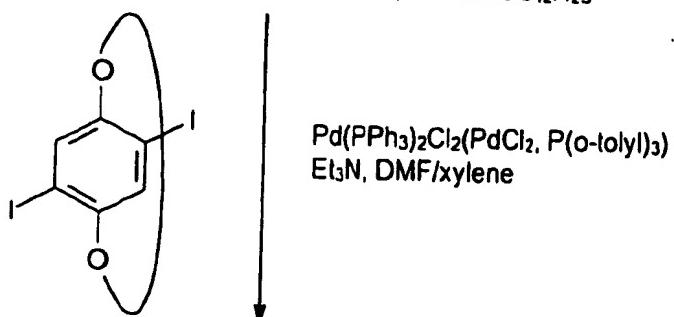
7b, R = C₁₂H₂₅

7c, R = COOC₁₂H₂₅

8a, R = OC₁₆H₃₃

8b, R = C₁₂H₂₅

7a, R = COOC₁₂H₂₅



9a, R = OC₁₆H₃₃

9b, R = C₁₂H₂₅

Scheme 2

These devices may be constructed in accordance with deposition and assembly techniques known in the art. The present invention may be used in the creation of a wide variety of lighting and lighted displays, giving the many advantages associated with polymeric materials.

In accordance with the present invention, results are presented for bilayer devices using PVK as a hole-transport layer and a family of copolymers of PPV and poly(pyridyl vinylene) PPyV with various side groups as the emitting layers. The absorption, photoluminescence and electroluminescence spectra indicate that the PL and EL are attributed to the formation of an exciplex at the PVK/copolymer interface for all the copolymer systems studied. An exciplex, like an excimer, is an excited state complex, except that an exciplex is formed between two different molecules (polymers in this case) rather than identical ones for an excimer [19]. Contrary to



expectations, earlier reported devices do not exhibit exciplex formation. For example, Greenham *et al* reported a bilayer device with CN-PPV and PPV, but the EL matches the PL and EL of a single CN-PPV film [3]. Results for other bilayer configurations also do not support exciplex formation [2]. Osaheni and Jenekhe [20] have observed exciplex formation in bilayers of PBO and TTA, but only for PL, although they do suggest that exciplexes may be important processes in organic light-emitting devices [20-21]. PL and EL due to exciplex formation has been reported in *blends* of PVK and a multiblock copolymer by Karasz and coworkers [17], but devices with separate layers were not investigated.

Brief Description of the Drawings

10 Figure 1 depicts three chemical structures relevant to the present invention: (a) a copolymer of PPyV and PPV with side groups R=C₁₂H₂₅ or COOC₁₂H₂₅, (b) a copolymer with side group R₁=OC₁₆H₃₃ and strap R₂=C₁₀H₂₀, and (c) a hole-transport layer poly(vinyl carbazole).

15 Figure 2 depicts plots showing absorbance of a single layer of PVK, a single layer of copolymer, and a bilayer of PVK/copolymer: (a) PPyVP(COOC₁₂H₂₅)₂V, (b) PPyVP(C₁₂H₂₅)₂V and (c) "strapped" copolymer.

20 Figure 3 shows photoluminescence of (a) PPyVP(C₁₂H₂₅)₂V film (dashed line), PVK/PPyVP(C₁₂H₂₅)₂V bilayer film (solid line), PVK film (O) (b) PPyVP(COOC₁₂H₂₅)₂V film (dashed line), PVK/PPyVP(COOC₁₂H₂₅)₂V bilayer film (solid line), PVK film (O), and (c) strapped copolymer film (dashed line), PVK/strapped copolymer bilayer film (solid line), PVK film (O).

Figure 4 shows electroluminescence (solid lines) for a (a) ITO/PVK/PPyVP($\text{COOC}_{12}\text{H}_{25}\right)_2\text{V}/\text{Al}$ device and a (b) ITO/PVK/strapped copolymer/Al device, and photoluminescence (dashed lines) for bilayer films of PVK and (a) PPyVP($\text{COOC}_{12}\text{H}_{25}\right)_2\text{V}$ and (b) strapped copolymer.

Figure 5 shows current density-voltage characteristics (\circ) and brightness-voltage characteristics of an ITO/PVK/PPyVP($\text{COOC}_{12}\text{H}_{25}\right)_2\text{V}/\text{Al}$ device. Inset shows a comparison between a single layer, ITO/PPyVP($\text{COOC}_{12}\text{H}_{25}\right)_2\text{V}/\text{Al}$, device (\square) and the bilayer device (\circ). The bilayer device is 10 times brighter at 10 times lower current density implying a 100 times improvement in efficiency.

Detailed Description of the Preferred Embodiment

In accordance with the foregoing summary of the invention, the following presents a detailed description of the preferred embodiment of the invention which is presently considered to be its best mode.

The synthesis of the PPyVPV copolymers is described elsewhere [22]. Figure 1(a) shows the molecular structure of poly(pyridyl vinylene phenylene vinylene) (PPyVPV). We report results for copolymers with side groups $\text{R}=\text{COOC}_{12}\text{H}_{25}$ and $\text{C}_{12}\text{H}_{25}$. Figure 1(b) shows the same copolymer with a "strap" across the phenyl ring in alternate PPV segments. For the second copolymer the side groups are $\text{R}_1=\text{OC}_{16}\text{H}_{33}$ with a strap $\text{R}_2=\text{C}_{10}\text{H}_{22}$. The copolymers are soluble in common organic solvents such as tetrahydrofuran (THF), xylene, and chloroform. The PVK (Fig. 1(c)) was purchased from Aldrich Chemical Co.

The PL and EL measurements were made using a PTI QM1 luminescence spectrometer. The absorption measurements were made using a Perkin Elmer Lambda 19 UV/Vis/NIR spectrometer. The current-voltage characteristics were measured using two Keithley 195A multimeters. The voltage was applied using a HP 6218A dc power supply. Quantum efficiency measurements were

made using a calibrated QTH lamp and a United Detector Technologies Silicon Photodiode (UV-100).

The samples for PL and absorption measurements were spin cast on quartz. PVK was spin cast (~3000 rpm) from a 10 mg/ml solution in THF. The copolymers were all spin cast (~1000 rpm) from xylene (~ 10 mg/ml) so the underlying PVK layer was not dissolved. For devices the polymers were spin cast on indium tin-oxide (ITO) substrates, which had previously been cleaned. The PVK layer in the bilayer configurations was dried for ~ 30 seconds in a laminar flow hood before the copolymer layer was spin cast. All fabrication steps for the devices were conducted in an ambient atmosphere in a Class 100 cleanroom. Aluminum electrodes were thermally evaporated onto the copolymer surface at low pressures ($< 10^{-6}$ torr). The active area of the devices was 7 mm².

Results and Discussion

Figure 2 shows the absorbance for single and bilayer systems. Each of the plots shows the absorbance of a single layer of PVK, a single layer of a copolymer, and a bilayer configuration of PVK and the corresponding copolymer. The onset of PVK absorption is at 3.5 eV and shows two spectral features at 3.6 and 3.75 eV similar to previous reports [6,7]. In each of the three cases (a), (b) and (c) of Fig. 2 the absorbance of the bilayer configuration is the sum of the absorbance of each of the individual layers. No new ground to excited state transitions are present. Photoluminescence excitation (PLE) results (not shown) for each of the systems confirm these results.

The photoluminescence for the single layer and bilayer configurations are shown in Fig. 3. The PL (excited at 3.6 eV) of a PVK film is shown in Fig. 3a, 3b and 3c by the open circles (O) and peaks at 3.05 eV similar to previous reports [6,7]. The dashed line is the PL of a single layer film of

each copolymer, (a) PPyVP($C_{12}H_{25}$)₂V, (b) PPyVP(COOC₁₂H₂₅)₂V and (c) the strapped copolymer. The spectra are similar with each of the peaks at ~ 2.1 eV with the exception of the strapped copolymer which also has a significant shoulder at 2.25 eV. The PL of the copolymer films, which peak near 2.6 eV [23], are significantly redshifted from that of the solution PL (not shown). The PL 5 redshift from solution to film is due to aggregation in the copolymer films [23].

The solid lines in Fig. 3 are the PL spectra for the bilayer configurations of PVK and each of the copolymers. In each case the bilayer films were excited at 3.6 eV an energy that is greater than the band gap of PVK. In each case, more prominently in Figs. 3b and 3c, there is PL emission at the same energy as the PVK PL emission (3.1 eV). However, the main feature in the PL of the bilayer 10 films is located at 2.5 eV for (a) PPyVP($C_{12}H_{25}$)₂V and (b) PPyVP(COOC₁₂H₂₅)₂V and at 2.4 eV for the (c) strapped copolymer. Emission at these energies is not observed for individual films of either PVK or the copolymers indicating that the emission is due to a completely different species, the exciplex. When the excitation energy is lowered below 3.4 eV (band gap of PVK) the emission due to the exciplex is drastically reduced. In addition, varying the concentration or thickness of the 15 copolymer or PVK films in the bilayer configuration will change the relative strengths of the exciplex peak and PVK peak.

PPyVP($C_{12}H_{25}$)₂V and PPyVP(COOC₁₂H₂₅)₂V have nearly identical PL results, which is expected since the side chains tend to perform the same function in both copolymers. The single layer PL results for the strapped copolymer in Fig. 3c show a completely new feature, a high energy 20 shoulder. The high energy shoulder is closer to the solution PL and is attributed to unaggregated sites in the film. The $C_{10}H_{20}$ strap around every other phenyl ring tends to disturb the aggregation that occurs in the other copolymers of this family. The same shoulder (now on the low energy side)

also appears in the bilayer film, indicating the PL has contributions from exciplex sites and from unaggregated regions of the strapped copolymer.

The bilayer devices have turn-on voltages ~ 12-16 volts with current densities between 0.1 and 0.5 mA/mm². The devices can easily be seen in a brightly lit room and have internal quantum efficiencies ~ 0.1-0.5%. Figure 5 shows the current-voltage (O) and voltage-brightness (solid line) characteristics for a typical ITO/PVK/PPyVP(COOC₁₂H₂₅)₂V/Al bilayer device. The inset of Fig. 5 shows a comparison between a single layer device (ITO/PPyVP(COOC₁₂H₂₅)₂V/Al) and the bilayer device shown in the main plot. The bilayer device is ten times brighter at an order of magnitude lower current density which means the bilayer device is ~ 100 times more efficient than the single layer device.

In the devices the electrons are injected from the Al electrode into the conduction band of the copolymer, but they are confined at the PVK/copolymer interface due to a large barrier. The holes injected from the ITO also may be confined at the interface by a somewhat smaller barrier. The increased number of electrons and holes in the interface region increase the probability of recombination via exciplex emission. In addition the buried interface severely reduces the non-radiative recombination that otherwise will occur near the electrodes.

Conclusion

In summary, the present invention demonstrates the presence of exciplex emission in heterojunctions of PVK and PPyVP(R)₂V. The addition of a C₁₀H₂₀ strap on every other phenyl ring in the copolymer reduces the aggregation in the films. Emission from the strapped copolymer bilayers is a combination of light from exciplex and unaggregated sites. The exciplex is the primary method of electroluminescence in the bilayer devices. The bilayer devices we have fabricated show

a 100 times increase in efficiency compared to single layer devices due to charge confinement and exciplex emission at the interface.

The following references are hereby incorporated herein by reference:

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The contents of U.S. Provisional Patent Application Serial Number 60/023,071 are hereby incorporated herein by reference.

In view of the present disclosure or through practice of the present invention, it will be within the ability of one of ordinary skill to make modifications to the present invention, such as through the use of equivalent arrangements and compositions, in order to practise the invention without departing from the spirit of the invention as reflected in the appended claims.

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(the next page is page 22)



In The Claims:

1. A light emitting polymeric material said light emitting polymeric material capable of producing electroluminescence upon being provided with a flow of electrons, said light emitting polymeric material comprising:

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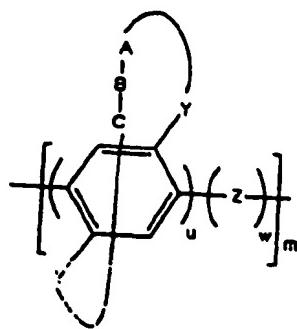
a plurality of polymeric chains comprising polymeric chains each having substituent moieties of sufficient number and size and extending from said polymeric chain and about a substantial portion of the circumference about said polymer chain so as to maintain said polymeric chains in a sufficiently deaggregated state, so as to substantially prevent the redshifting of said

10 electroluminescence and the lowering of light emission efficiency of said electroluminescence.

2. A light emitting polymeric material according to claim 1 comprising polymeric chains selected from the group consisting of alternating and random copolymers, having the structure:

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wherein m is the degree of polymerization;

Y is selected from the group consisting of - CH_2 , O , S , CO and NR_2 wherein R is an alkyl group containing 1 to 16 carbon atoms;

5 A is selected from the group consisting of $(\text{CH}_2)_n$, $(\text{CH}_2\text{CH}_2\text{O})_n$, $(\text{CH}_2\text{CH}_2\text{O})_n\text{NR}$; wherein R is an alkyl group containing 1 to 16 carbon atoms, and aryl groups having 6 to 14 carbon atoms;

B is selected from the group consisting of $(\text{CH}_2)_n$, aryl groups having 6 to 14 carbon atoms, and calixarene having 18 to 200 carbon atoms;

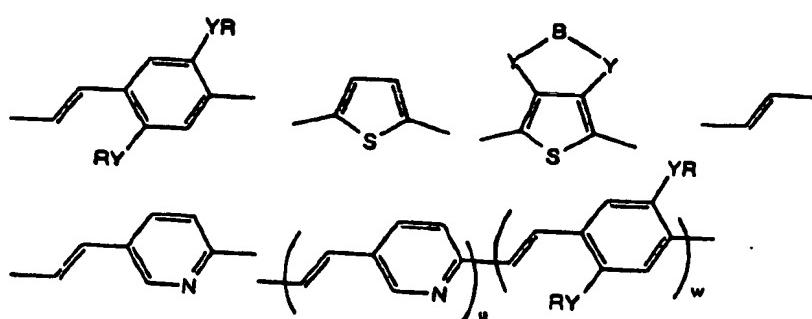
wherein u may be of a value independently selected from the group 1 to 6, inclusive;

10 wherein w may be of a value independently selected from the group 1 to 6, inclusive;

wherein n may be of a value independently selected from the group 0 to 6, inclusive; and

wherein Z may be a structure selected from the group consisting of :

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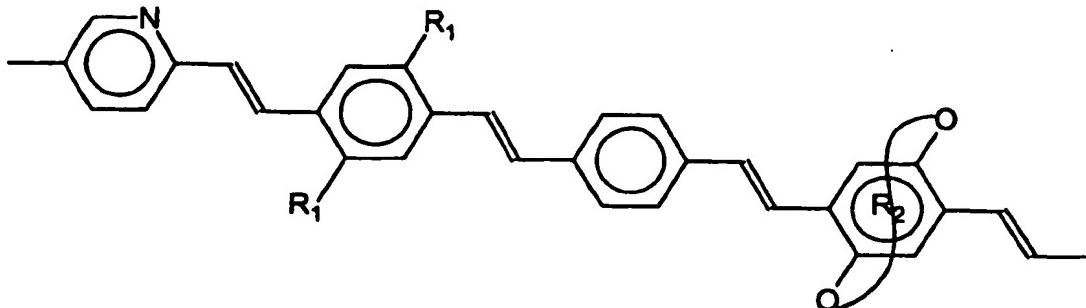
wherein R is an alkyl group containing 1 to 16 carbon atoms;

20 wherein Y is selected from the group consisting of - CH_2 , O , S , CO and NR_2 wherein R is an alkyl group containing 1 to 16 carbon atoms;

B is selected from the group consisting of $(CH_2)_n$, aryl groups having 6 to 14 carbon atoms, and calixarene having 18 to 200 carbon atoms;

wherein u may be of a value independently selected from the group 1 to 6, inclusive; and wherein w may be of a value independently selected from the group 1 to 6, inclusive.

- 5 3. A light emitting polymeric material according to claim 1 wherein said polymer comprises polymeric chains selected from the group consisting of copolymers, having the structure:



wherein R₁ is OC₁₆H₃₃ and R₂ is C₁₀H₂₀.

- 10 4. A light emitting polymeric material according to any one of claims 1 to 3 wherein said polymeric material is further provided with a layer of an electron blocking polymer.
- 15 5. A light emitting polymeric material according to claim 4 wherein said electron blocking polymer is selected from the group consisting of poly(vinylcarbazole).
- 20 6. A light emitting polymeric material said light emitting polymeric material capable of producing electroluminescence upon being provided with a flow of electrons, said light emitting polymeric material comprising:
a plurality of polymeric chains comprising polymeric chains each being provided with rotaxanes of sufficient number and size and extending from said polymeric chain and about a substantial portion of the circumference about said polymer chain so as to maintain said polymeric chains in a sufficiently deaggregated state, so as to substantially prevent the redshifting of said electroluminescence and the lowering of light emission efficiency of said electroluminescence.
- 25 7. A light emitting polymeric material according to claim 6 wherein said polymeric material is further provided with a layer of an electron blocking polymer.
8. A light emitting polymeric material according to claim 7 wherein said electron blocking polymer is selected from the group consisting of poly(vinylcarbazole).
9. A light emitting polymeric material, substantially as hereinbefore described with reference to any one of the examples.



10. A process for producing a light emitting polymeric material, said process substantially as hereinbefore described with reference to Scheme 1 and Scheme 2.

11. A light emitting polymeric material produced by the process of claim 10.

12. A light emitting device, said device comprising a light emitting polymeric material according to any one of claims 1 to 9 and 11, and a source of electrical current so as to supply said electron transporting polymer with a flow of electrons.

13. A light emitting device, said device comprising a light emitting polymeric material according to any one of claims 1 to 9 and 11, and a source of electrical current so as to supply said electron transporting polymer with a flow of electrons said device selected from the group consisting of single layer, bilayer and multi-layer light emitting devices.

14. A light emitting device, said device comprising a light emitting polymeric material, substantially as hereinbefore described with reference to any one of the examples.

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The Ohio State University Research Foundation

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